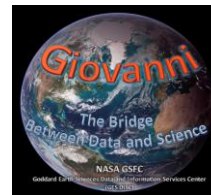# Characterizing the Photochemical Environment over China
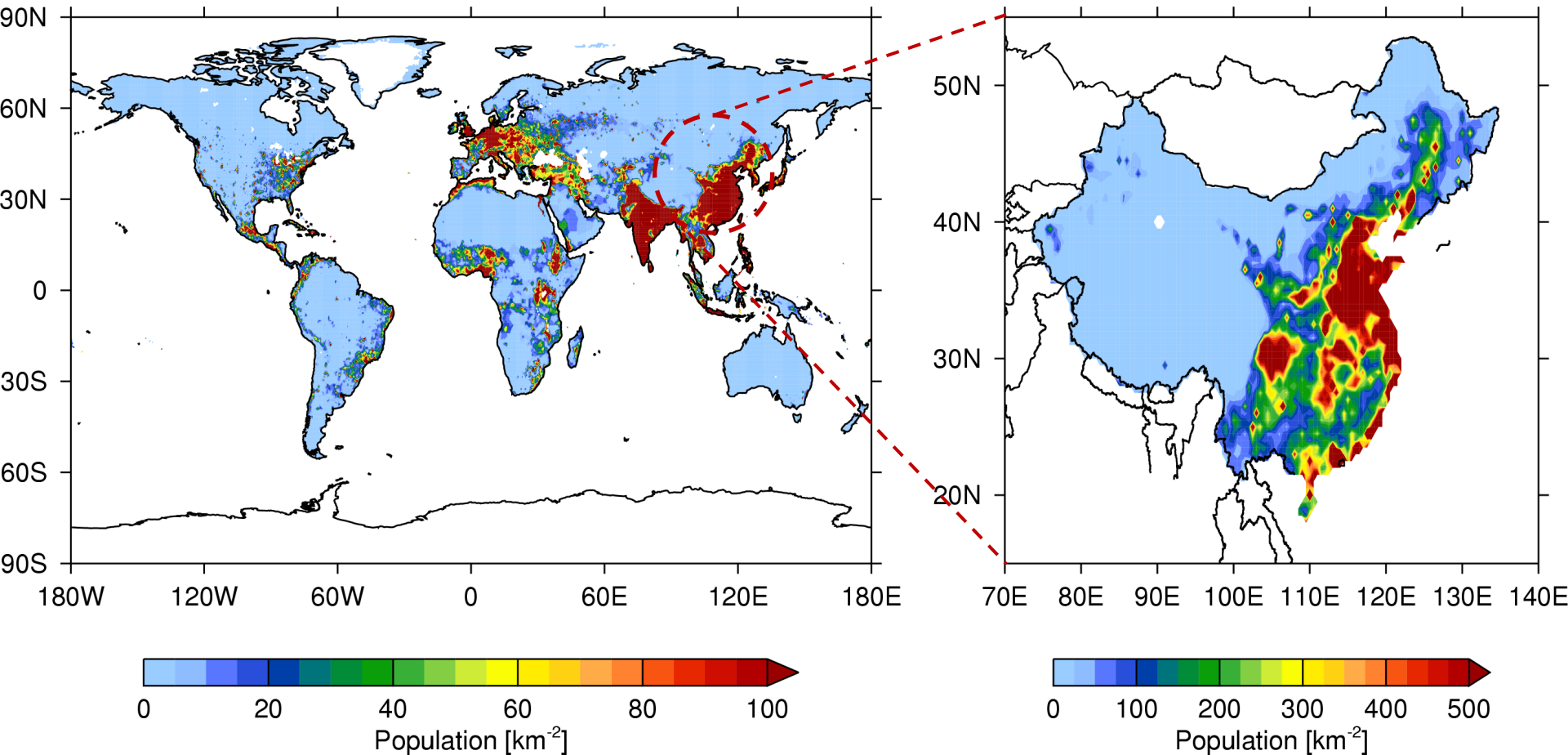
Zhen Liu

School of Earth and Atmospheric Sciences
Georgia Institute of Technology
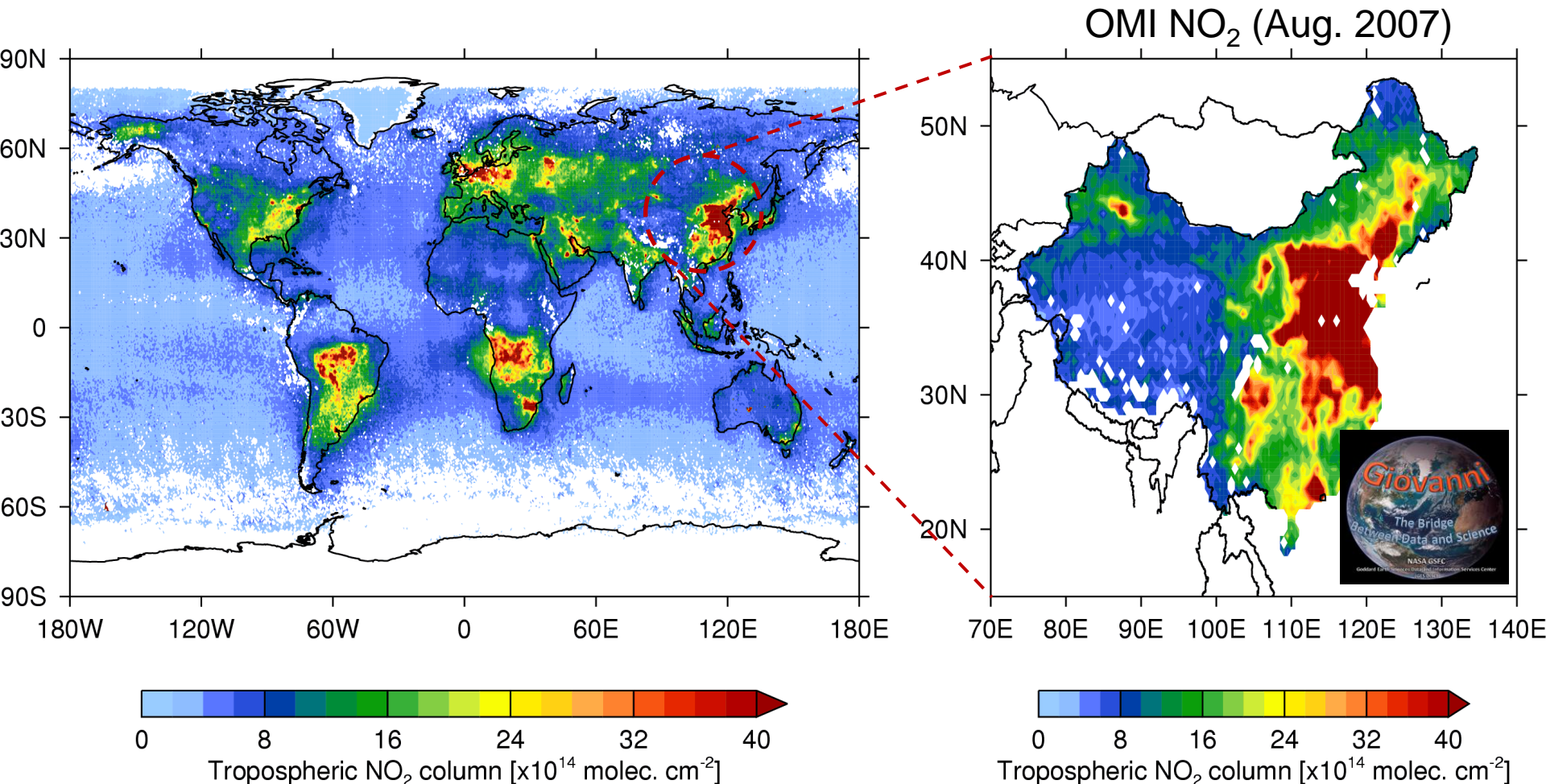
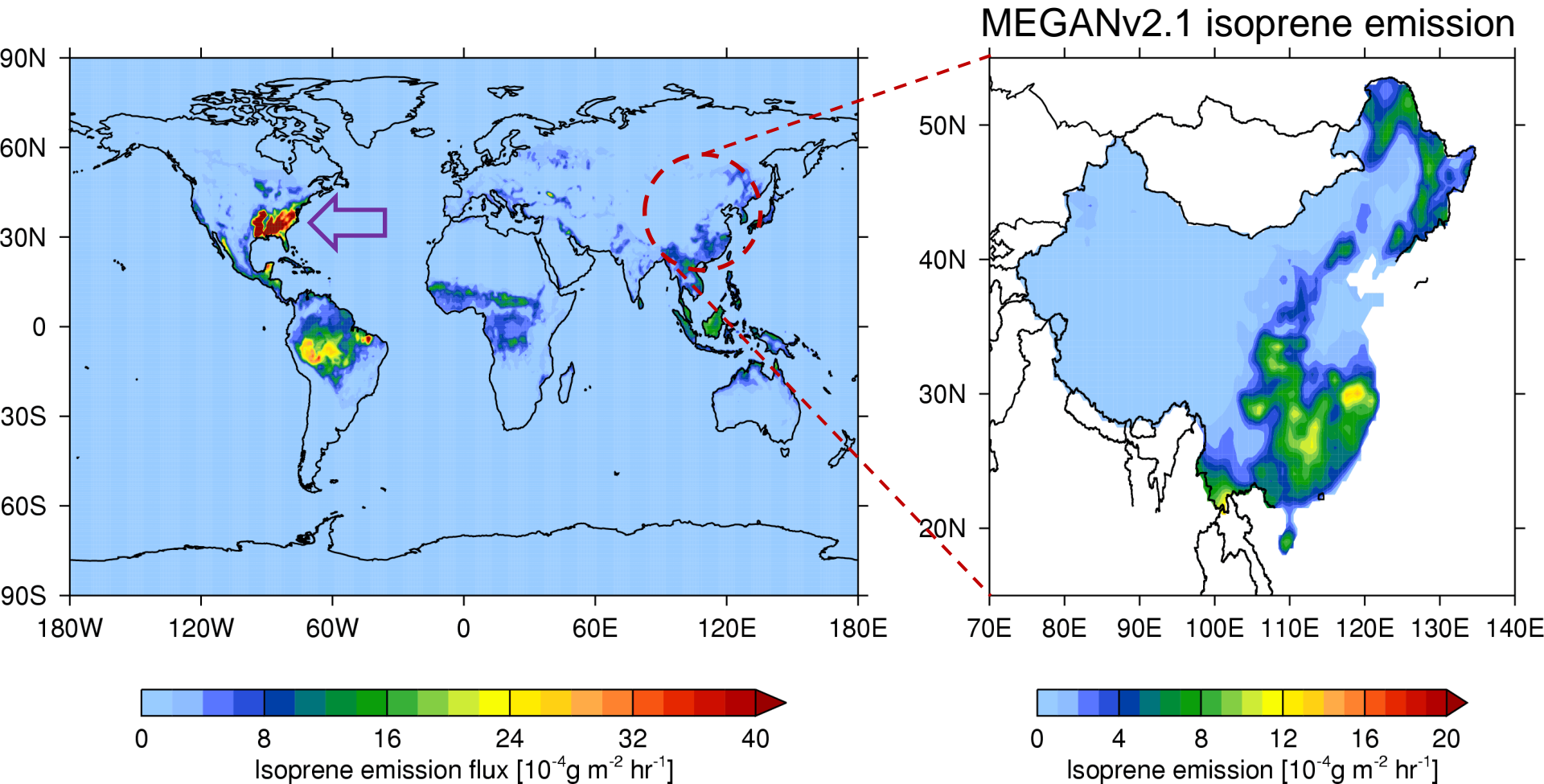26 September 2012

The region of concern - China

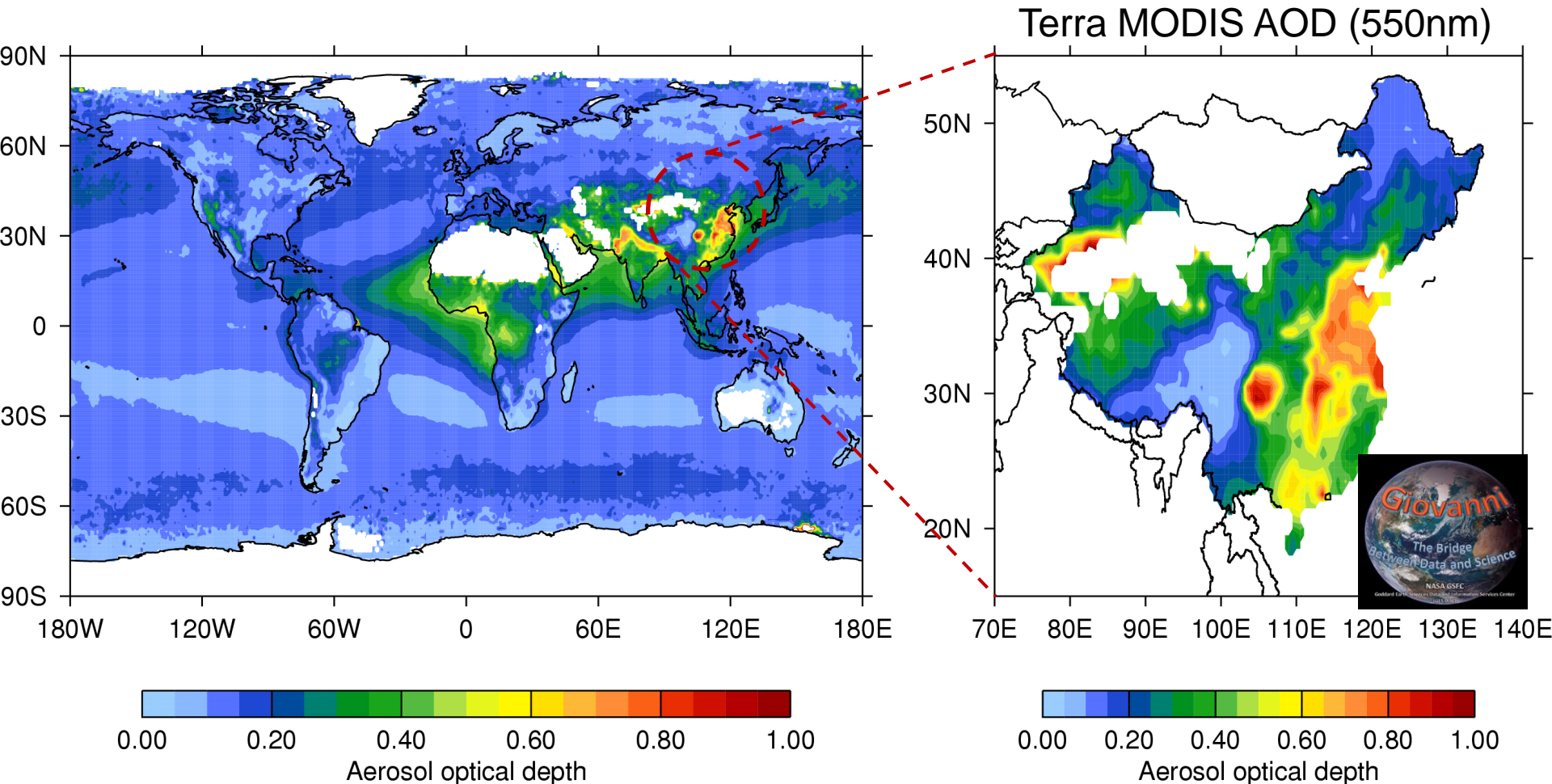The country with the largest population

The region of concern - China

OMI NO$_2$ (Aug. 2007)

Tropospheric NO$_2$ column [x10^{14} molec. cm^{-2}]

Tropospheric NO$_2$ column [x10^{14} molec. cm^{-2}]

Concentrated high anthropogenic emissions

MEGANv2.1 isoprene emission

Isoprene emission flux [10^{-4}g m^{-2} hr^{-1}]

Isoprene emission [10^{-4}g m^{-2} hr^{-1}]

Low biogenic emissions over polluted regions

The region of concern - China

Terra MODIS AOD (550nm)

Aerosol optical depth

The region with the highest aerosol abundance

Scientific Goal

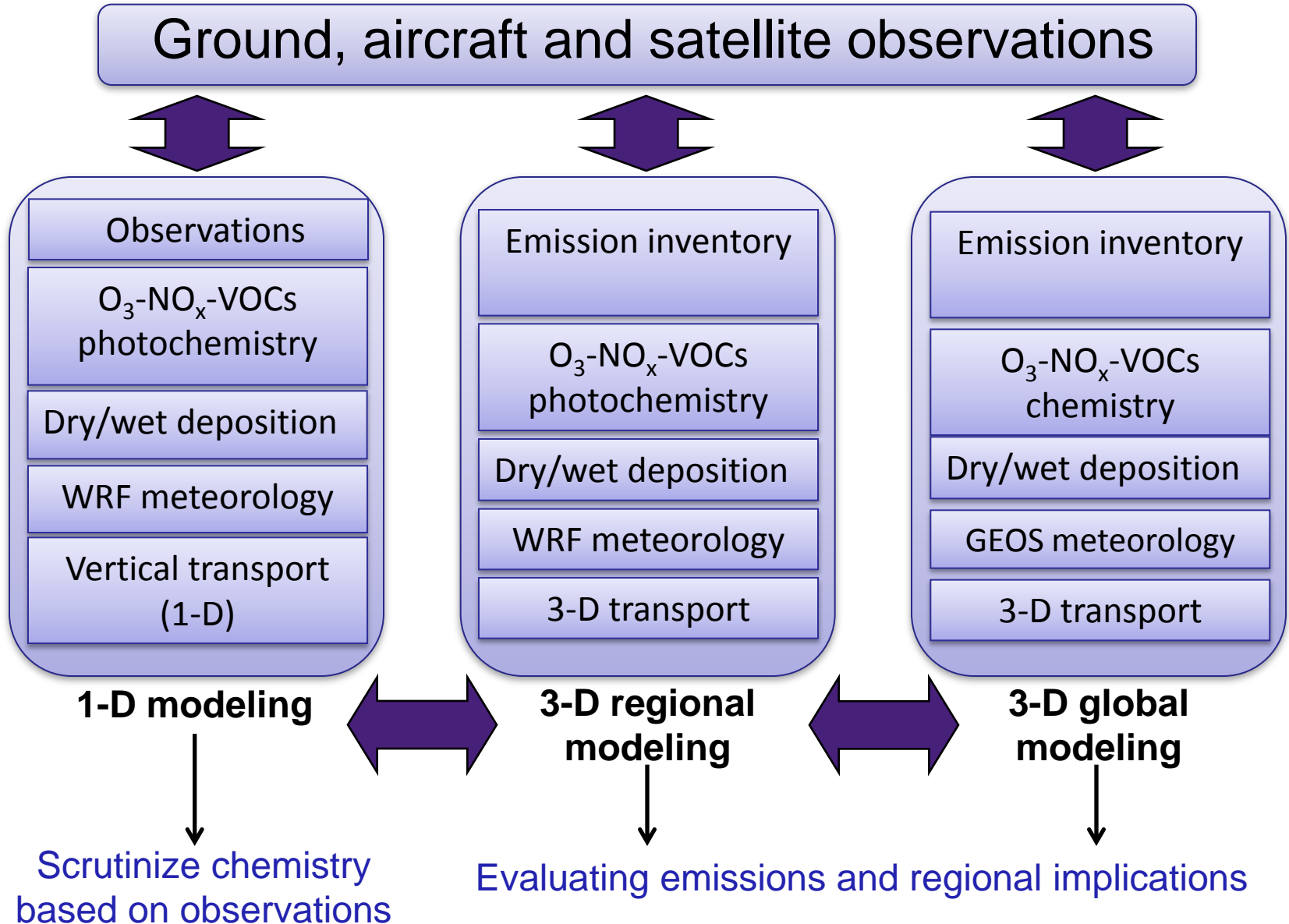| High anthropogenic emissions (e.g. NOx, VOCs) | **+** | Low biogenic emissions (e.g. isoprene) | **+** | High loading of aerosols |
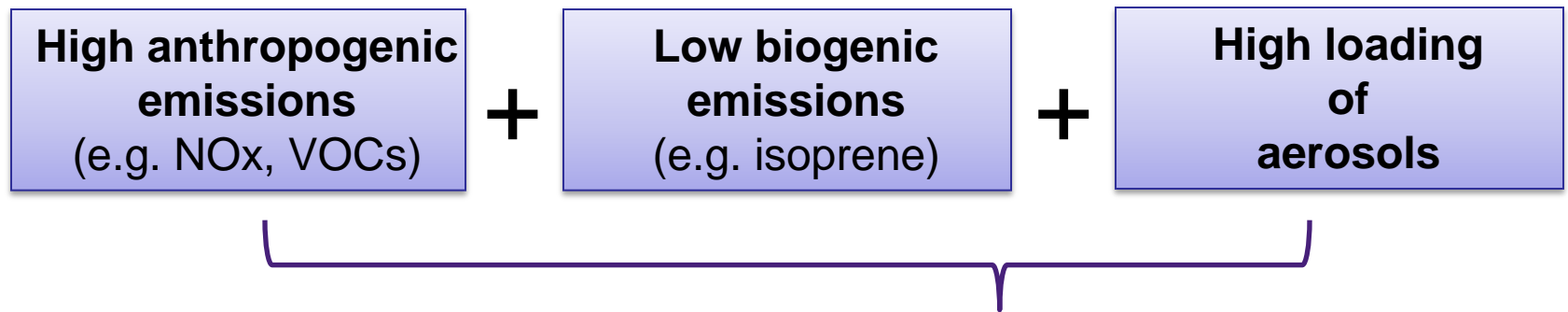
Characterize the **Photochemical Environment** –
To examine the processes (**emissions**, **chemistry**, **transport**) that regulate the observed chemical concentrations.

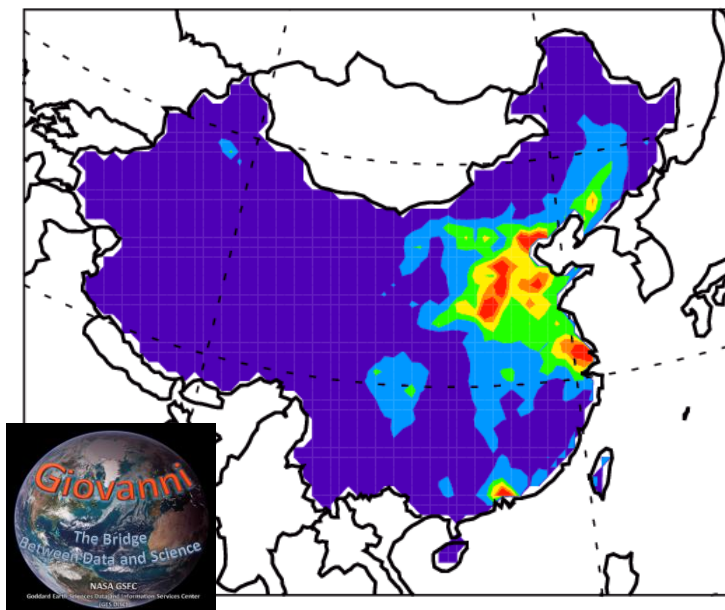$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\boldsymbol{u} c_i) = R_i(c_1, c_2, ..., c_n) + E_i - S_i$$

transport   **chemistry**   **emission**   deposition

Multi-scale modeling analysis

Ground, aircraft and satellite observations

1-D modeling

- Observations
- O_3-NO_x-VOCs photochemistry
- Dry/wet deposition
- WRF meteorology
- Vertical transport (1-D)

Scrutinize chemistry based on observations

3-D regional modeling

- Emission inventory
- O_3-NO_x-VOCs photochemistry
- Dry/wet deposition
- WRF meteorology
- 3-D transport

Evaluating emissions and regional implications

3-D global modeling

- Emission inventory
- O_3-NO_x-VOCs chemistry
- Dry/wet deposition
- GEOS meteorology
- 3-D transport

REAM-3D model evaluation – NO$_2$

OMI (August 2007)

REAM (70km, WRF driven)
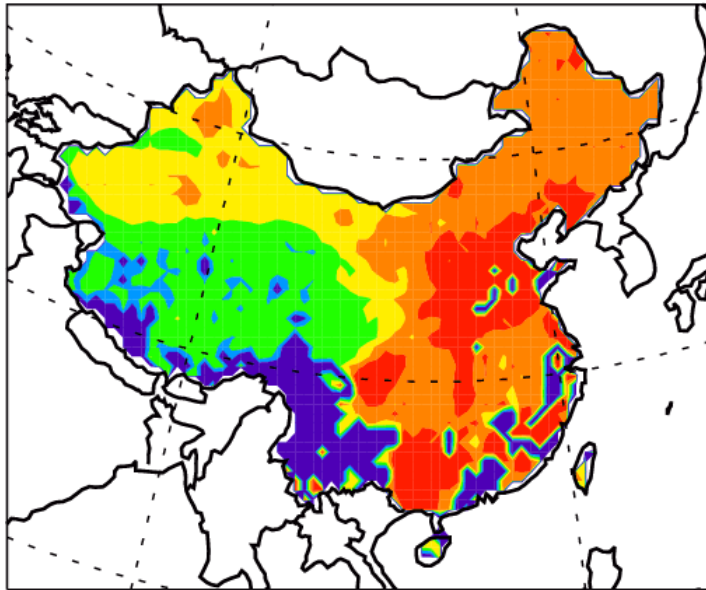w/inverted NO$_x$ emission by Zhao and Wang [2009]
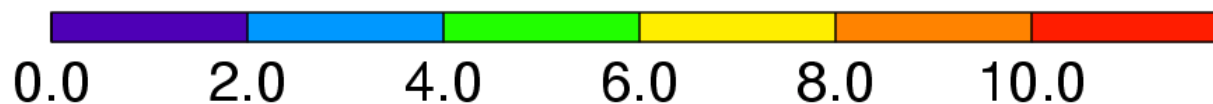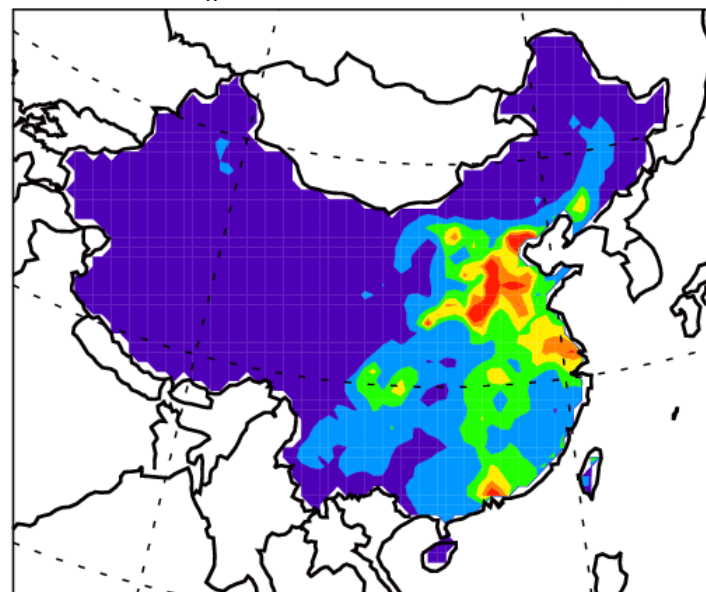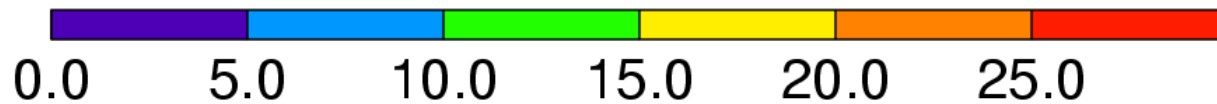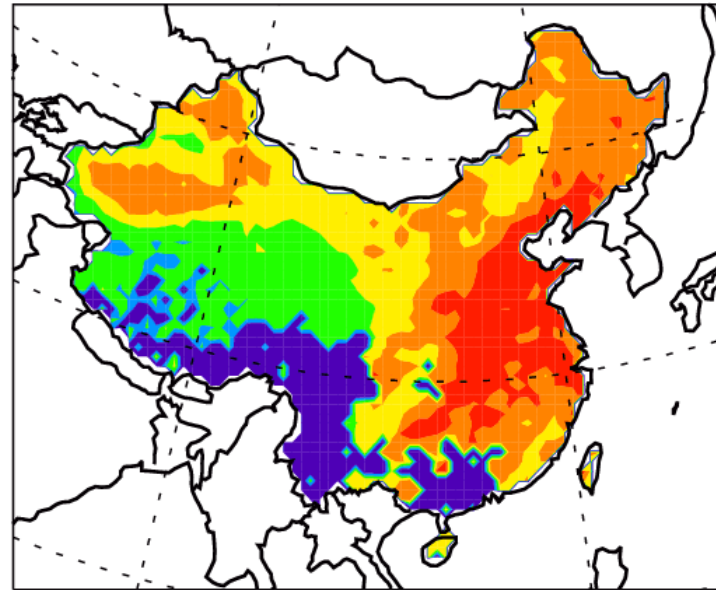

NO$_2$ [10^{15} molec cm^{-2}]

"Wow~ The model is perfect!"

REAM-3D model evaluation – CO

MOPITT (August 2007)

REAM (70km, WRF driven)
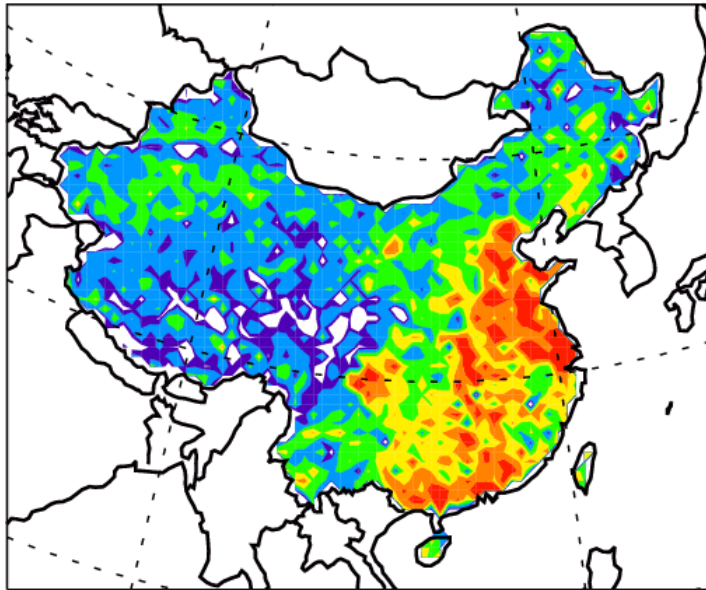w/ Streets 2006 inventory [Zhang et al., 2009]
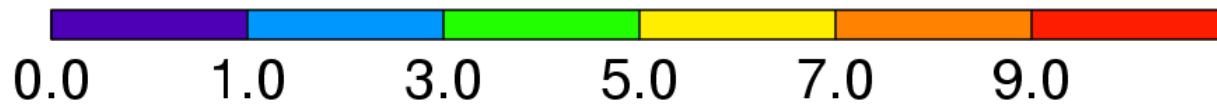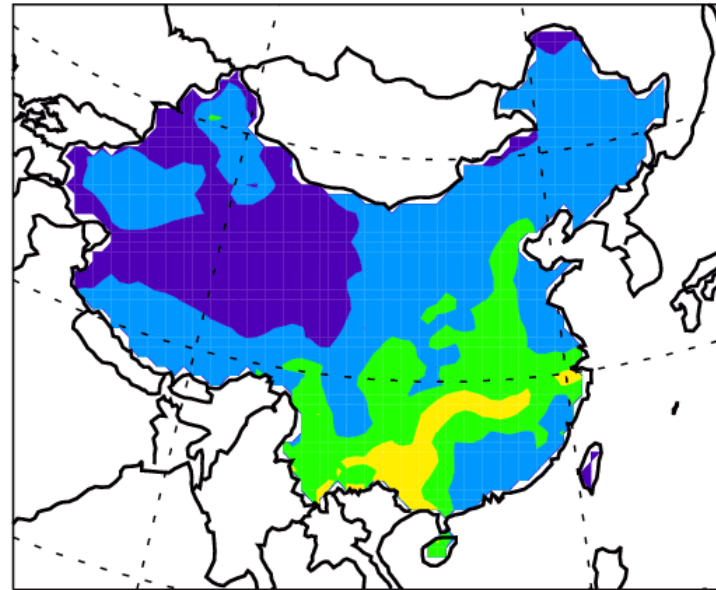
$CO \; [10^{17} molec \; cm^{-2}]$

"Not bad!"

REAM-3D model evaluation – CHOCHO

SCIAMACHY (August 2007)

REAM (70km, WRF driven)
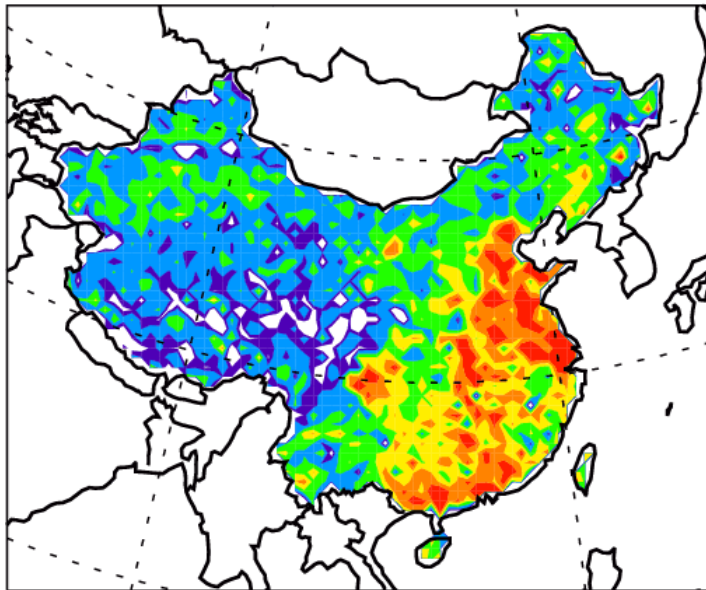w/ Streets 2006 VOC inventory [Zhang et al., 2009]

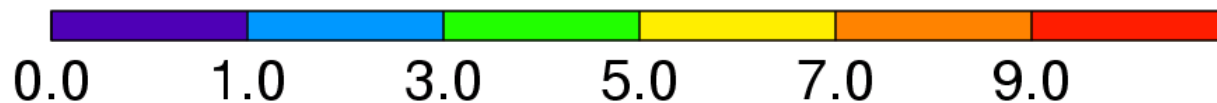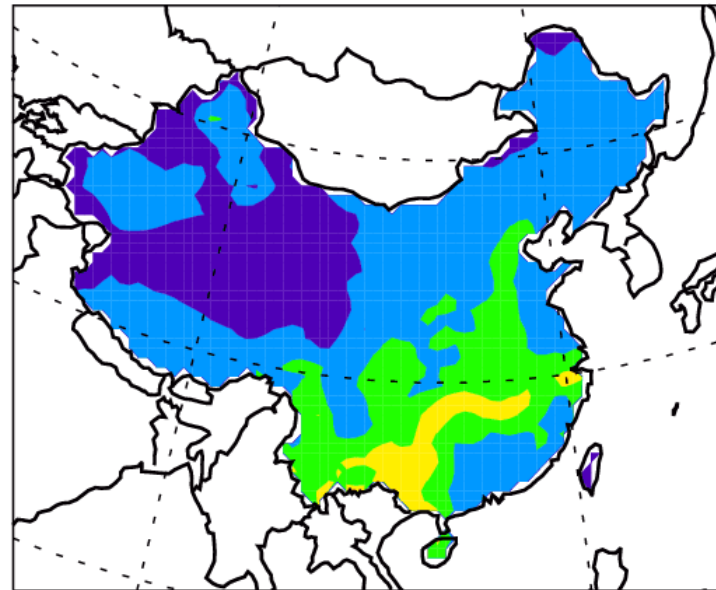CHOCHO [10^{14} molec cm^{-2}]

0.0   1.0   3.0   5.0   7.0   9.0

"What happened to the satellite?!"

REAM-3D model evaluation – CHOCHO

SCIAMACHY (August 2007)

REAM (70km, WRF driven)
w/ Streets 2006 VOC inventory [Zhang et al., 2009]

CHOCHO [10^{14} molec cm^{-2}]

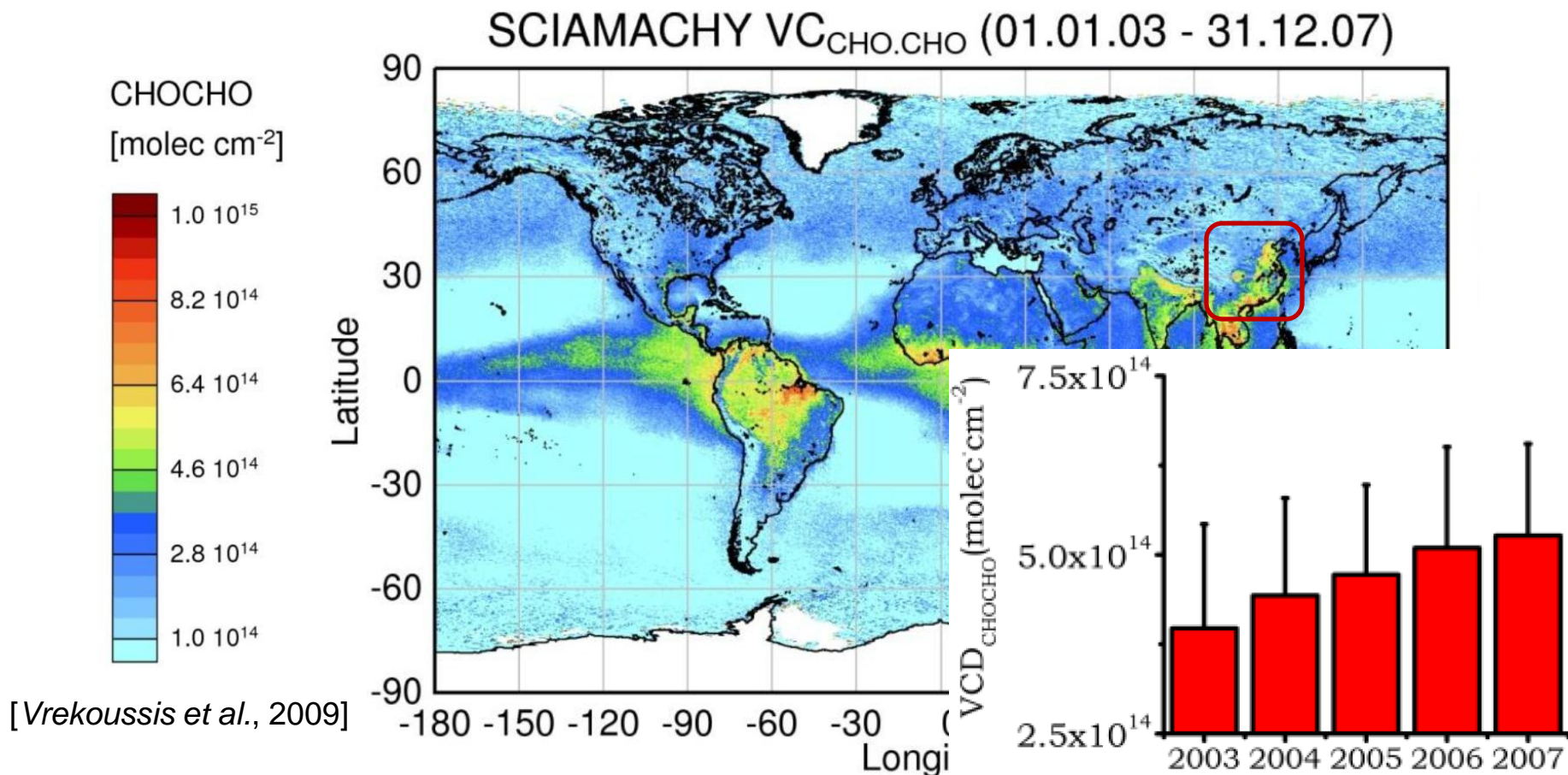Consistent with global modeling results of glyoxal budgets [e.g. Fu et al., 2008]

What is the missing source of glyoxal over China?

What is the missing source of glyoxal?

SCIAMACHY

SCIAMACHY VC$_{CHO.CHO}$ (01.01.03 - 31.12.07)

CHOCHO [molec cm^{-2}]
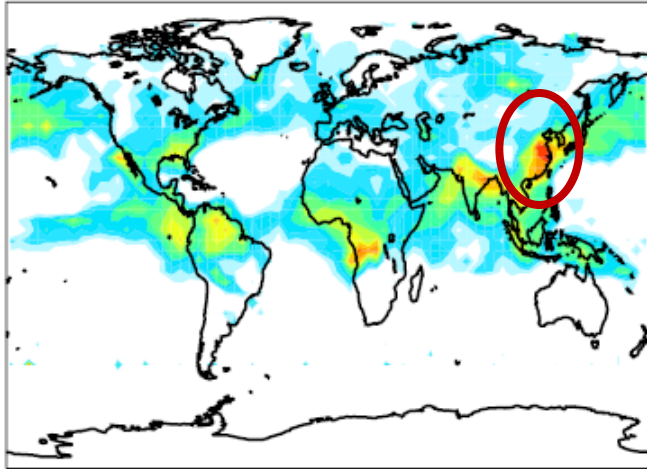
[*Vrekoussis et al.*, 2009]

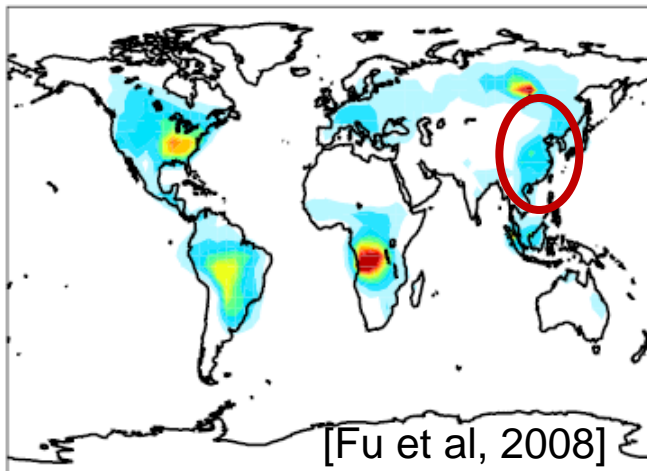A VOC oxidation product with minor primary emissions [*e.g. Volkamer et al.,* 2005]
Providing constraint for VOC chemistry and emissions

A missing source of CHOCHO over China

SCIAMACHY

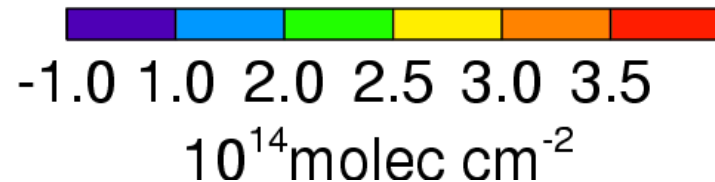**GEOS-Chem**

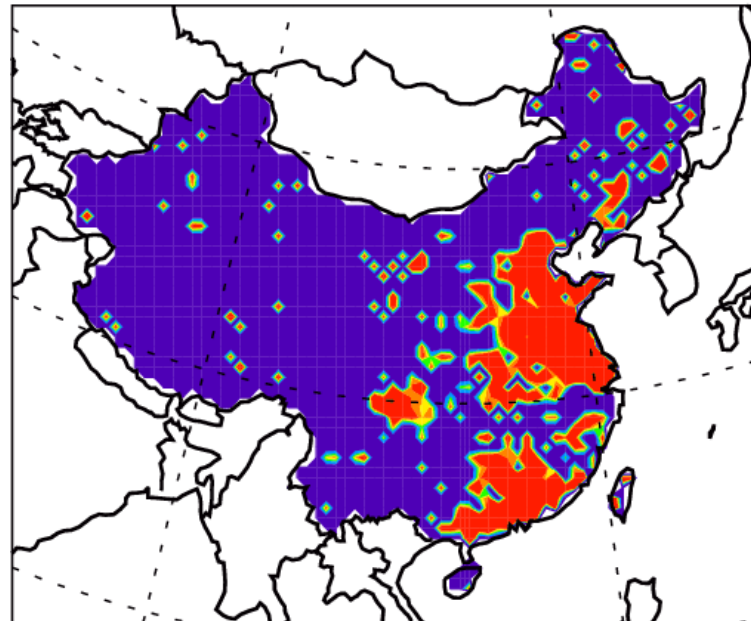[Fu et al, 2008]

10^{14} molec cm^{-2}

REAM CTM:
70km; WRF driven; Streets 2006; GFEDv2; MEGANv2.1; Explicit CHOCHO gas-phase chemistry; sampling at SCIAMACHY overpassing time (10am LT)
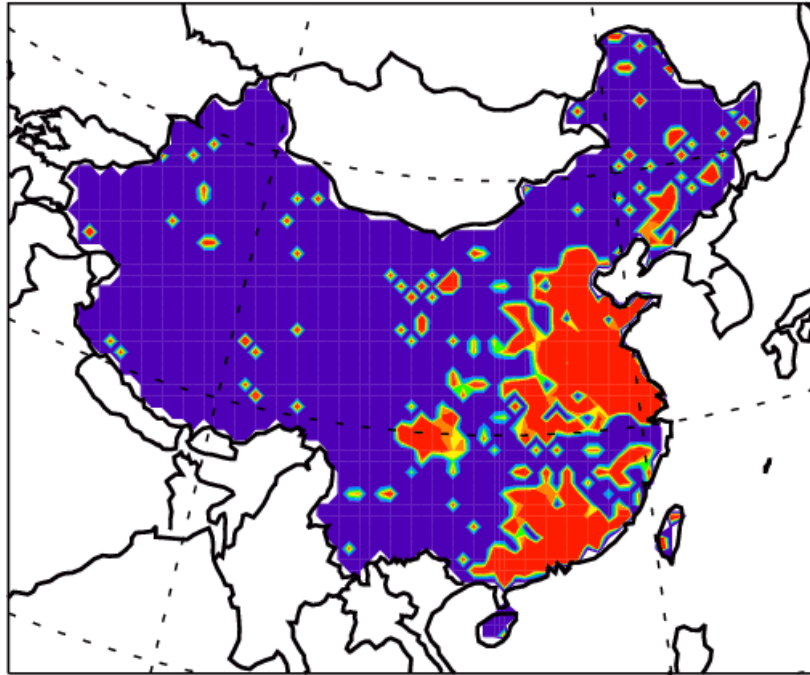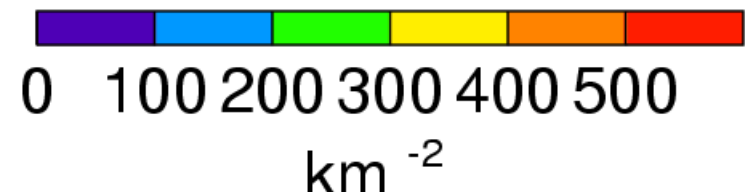
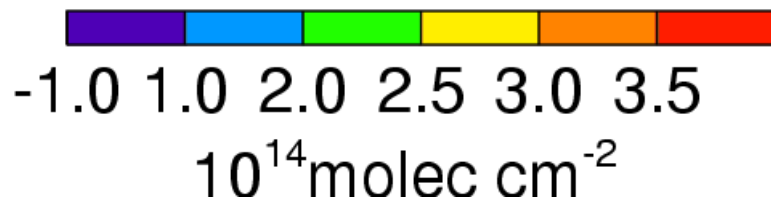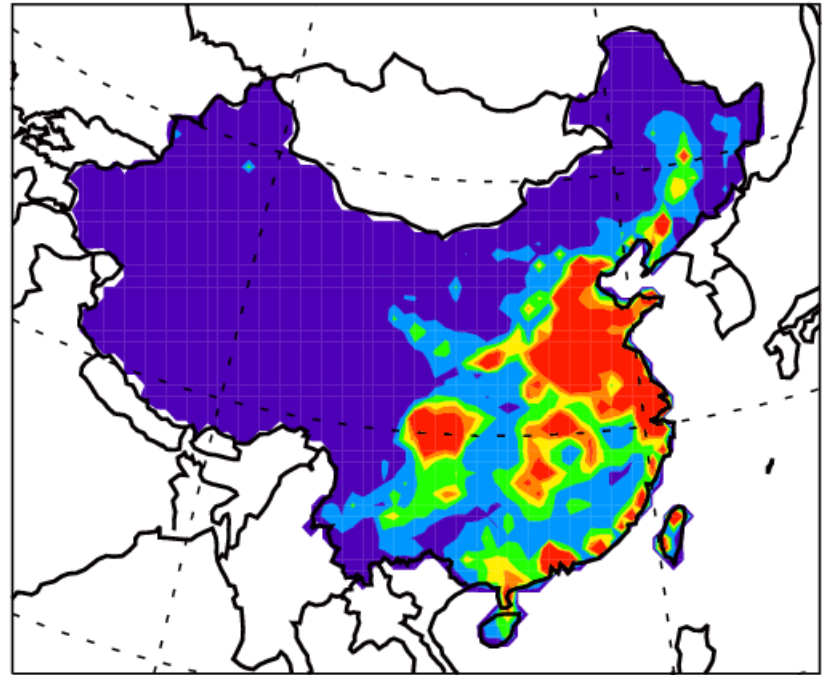$\triangle$**CHOCHO**
SCIAMACHY – REAM (Aug. 2007)

-1.0 1.0 2.0 2.5 3.0 3.5
10^{14}molec cm^{-2}

ΔCHOCHO$_{SCIA-REAM}$

Population density

-1.0 1.0 2.0 2.5 3.0 3.5
10^{14}molec cm^{-2}

0 100 200 300 400 500
km^{-2}

A missing source that is anthropogenic?

A Linearized Model

$$C_{CHOCHO} \approx \sum_{i=1}^{n} E_i \times \frac{\partial C_{CHOCHO}}{\partial E_i} + C_{CHOCHO}^0$$
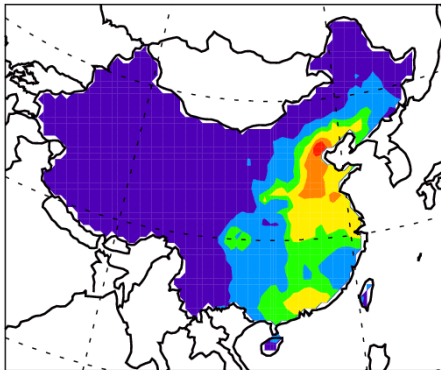
i: the *i* th of the *n* precursors;
E_i: Emission of the *i* th precursor
[*Fu et al., 2007*]

❑ Mostly over emission regions due to short lifetime
❑ Emissions are very uncertain

❑ Most reactive precursor contributing the most
❑ Significant only over southern China

❑ Creating a regional background of glyxoal
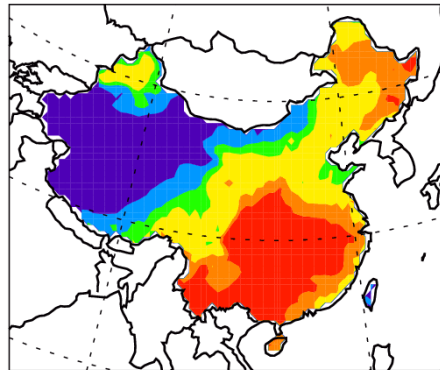❑ Reasonable emissions
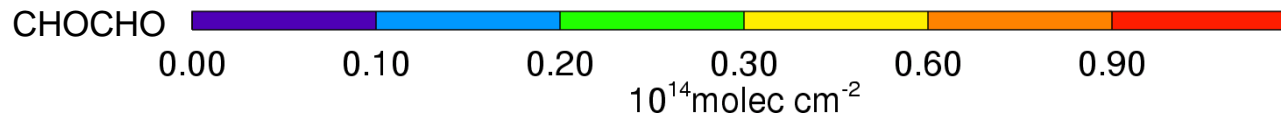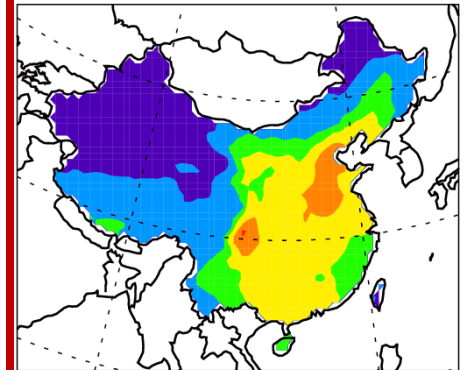[e.g. *Carmichael et al.*, 2003]
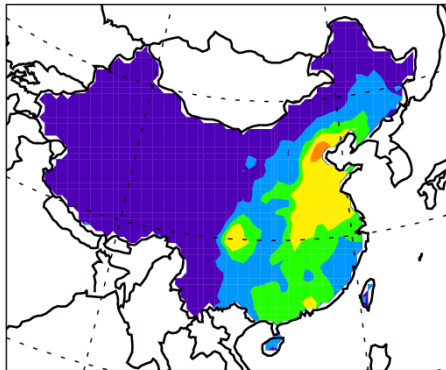
from ARO1 (E_1) from ARO2 (E_2) from ISOP (E_3) C_{CHOCHO}^0 from C_2H_2 and others

CHOCHO
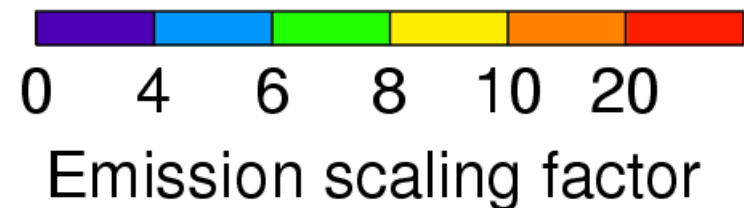
0.00   0.10   0.20   0.30   0.60   0.90
10^{14} molec cm^{-2}

Translate ∆CHOCHO into precursor emissions

$$\Delta C'_{CHOCHO} \approx \sum_{i=1}^{n} \Delta E_i \times \frac{\partial C_{CHOCHO}}{\partial E_i} \quad \Rightarrow \quad \Delta E_i \approx \frac{\Delta C'_{CHOCHO}}{\partial C_{CHOCHO} / \partial E_i} \quad \Rightarrow \quad S_i = \frac{\Delta E_i + E_i}{E_i}$$

∆CHOCHO$_{SCIA-REAM}$ 　　　　　　　　　ISOP

10^{14}molec cm^{-2}

Emission scaling factor

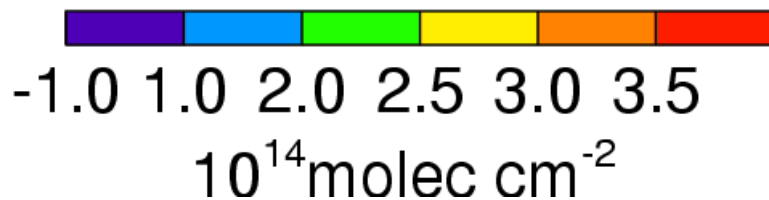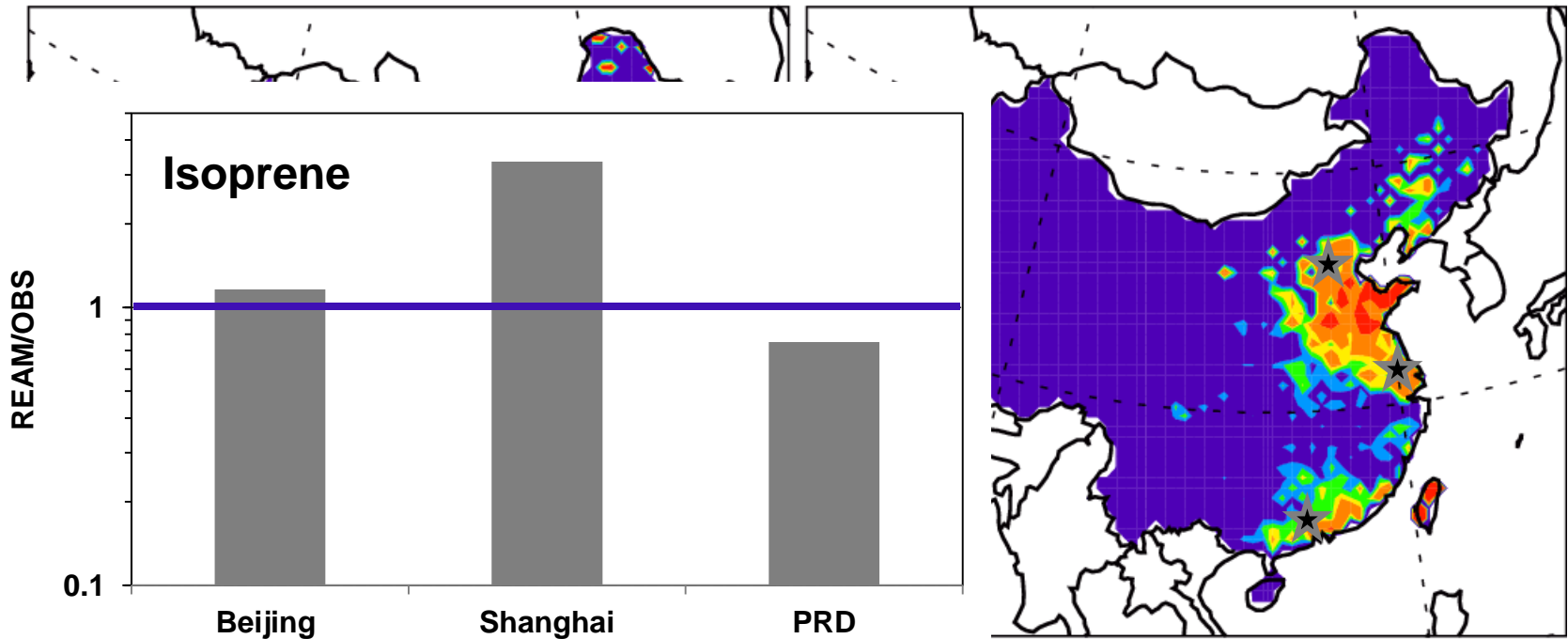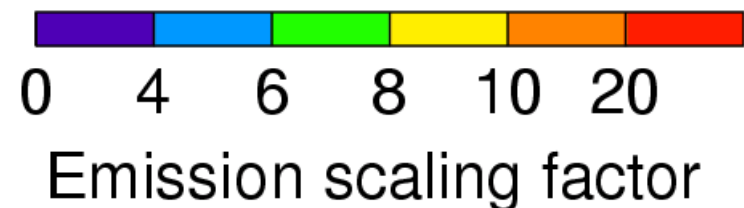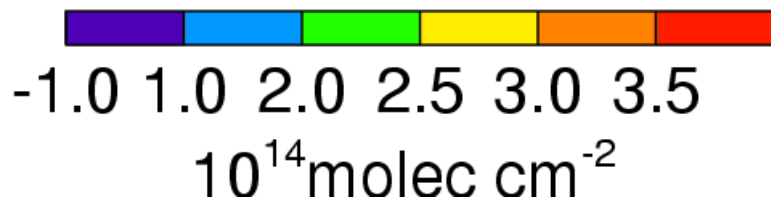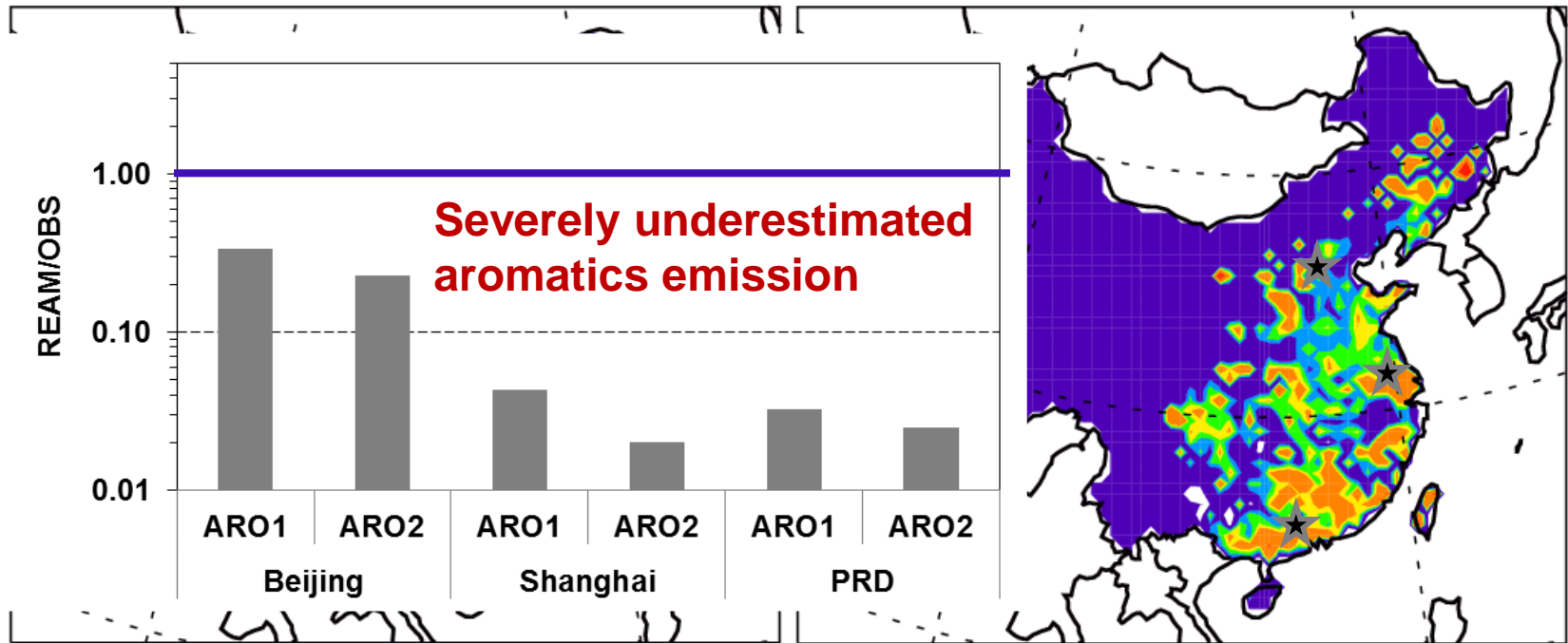# Translate ∆CHOCHO into precursor emissions

$$\Delta C'_{CHOCHO} \approx \sum_{i=1}^{n} \Delta E_i \times \frac{\partial C_{CHOCHO}}{\partial E_i} \implies \Delta E_i \approx \frac{\Delta C'_{CHOCHO}}{\partial C_{CHOCHO}/\partial E_i} \implies S_i = \frac{\Delta E_i + E_i}{E_i}$$

∆CHOCHO$_{SCIA-REAM}$

ARO1 + ARO2

Severely underestimated aromatics emission

REAM/OBS

1.00

0.10

0.01

ARO1	ARO2	ARO1	ARO2	ARO1	ARO2
Beijing		Shanghai		PRD	

-1.0 1.0 2.0 2.5 3.0 3.5
10^{14} molec cm^{-2}

0 4 6 8 10 20
Emission scaling factor

Model results with top-down emissions
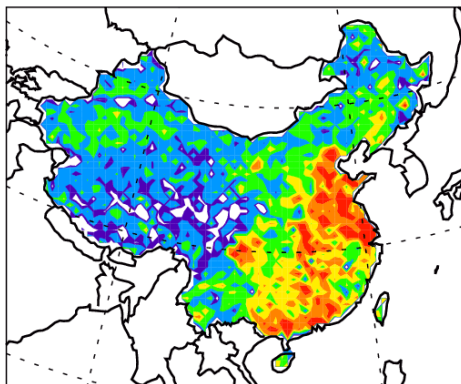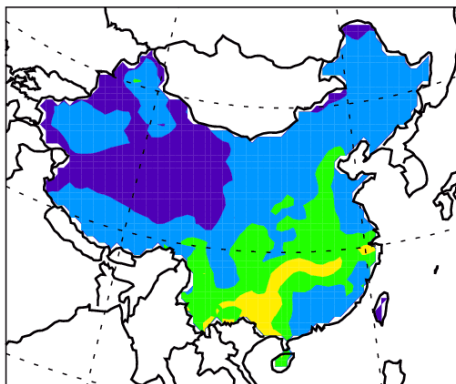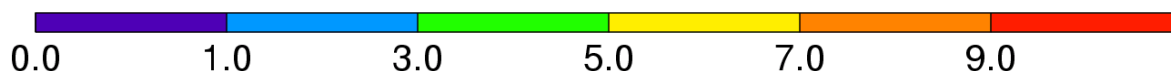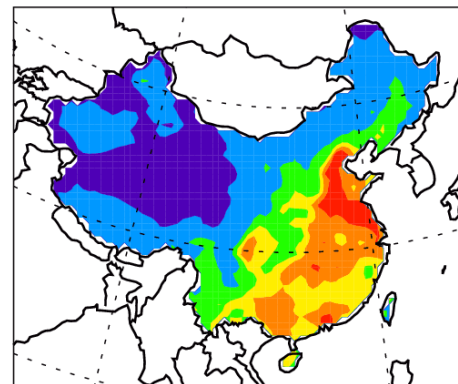

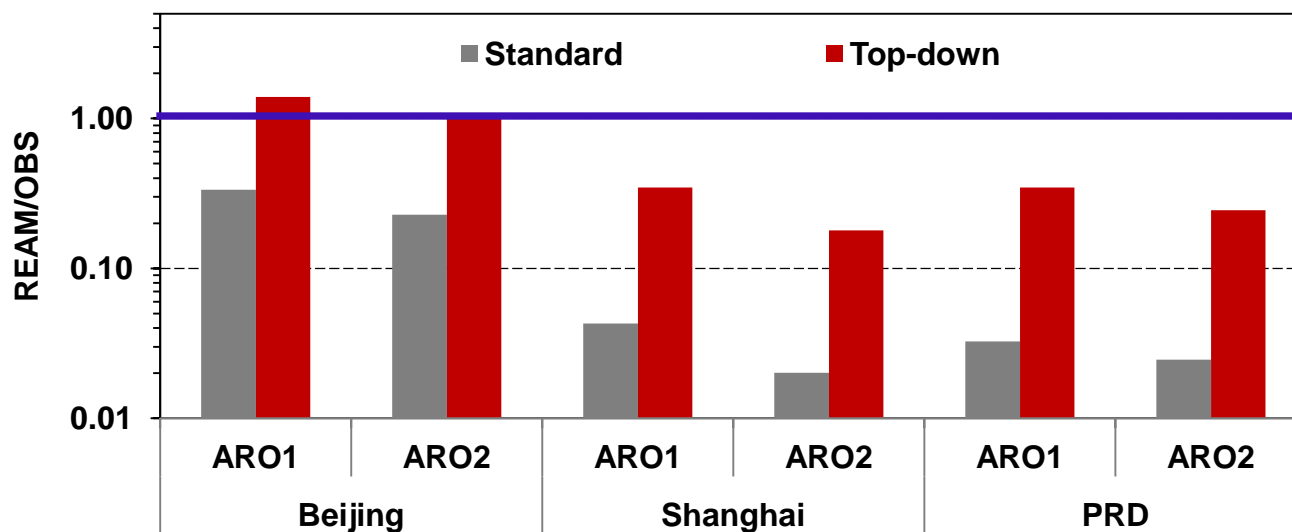(a) SCIAMACHY (b) REAM standard ARO (c) REAM top-down ARO
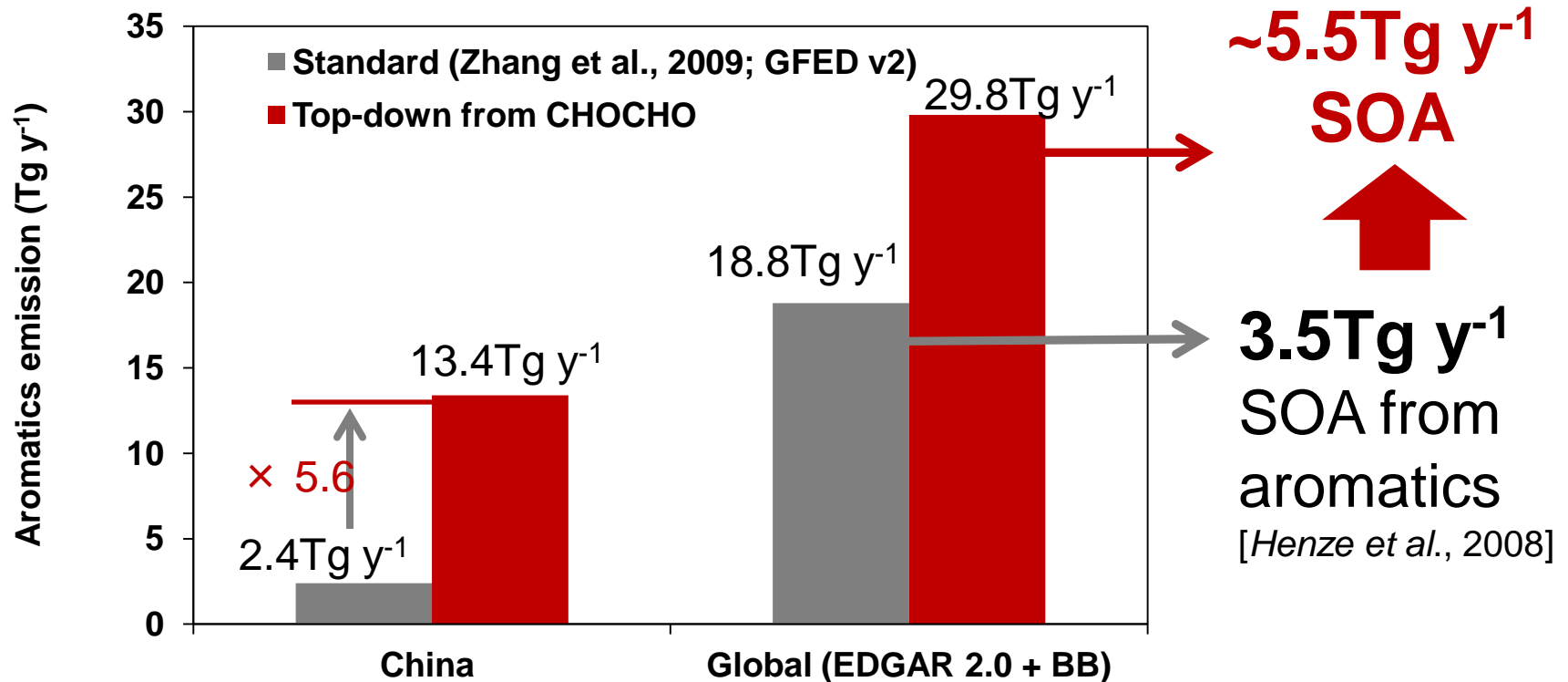
CHOCHO [10^{14} molec cm^{-2}]

Better agreement with observed aromatics at multiple sites;

Other than the impacts on PAN and O$_3$

How much more SOA from aromatic VOCs?

Partly explains previous underestimation of OA over the region
[*e.g. Heald, et al.,* 2005; *Fu et al., 2011,* ACPD]

Acknowledgement

My coauthors:

Yuhang Wang (my Ph.D advisor)
School of Earth and Atmospheric Sciences, Georgia Institute of Technology, USA

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College of Environmental Sciences and Engineering, Peking University, Beijing, China
Chih-Chung Chang, Shaw-Chen Liu
Research Center for Environmental Changes, Academic Sinica, Taipei, Taiwan
Hongli Wang, Changhong Chen
Shanghai Atmospheric Environment Institute, Shanghai Academy of Environmental Sciences, Shanghai, China

Thank Jim for having me in this workshop!!
Look forward to more and closer collaborations
with the GIOVANNI team!